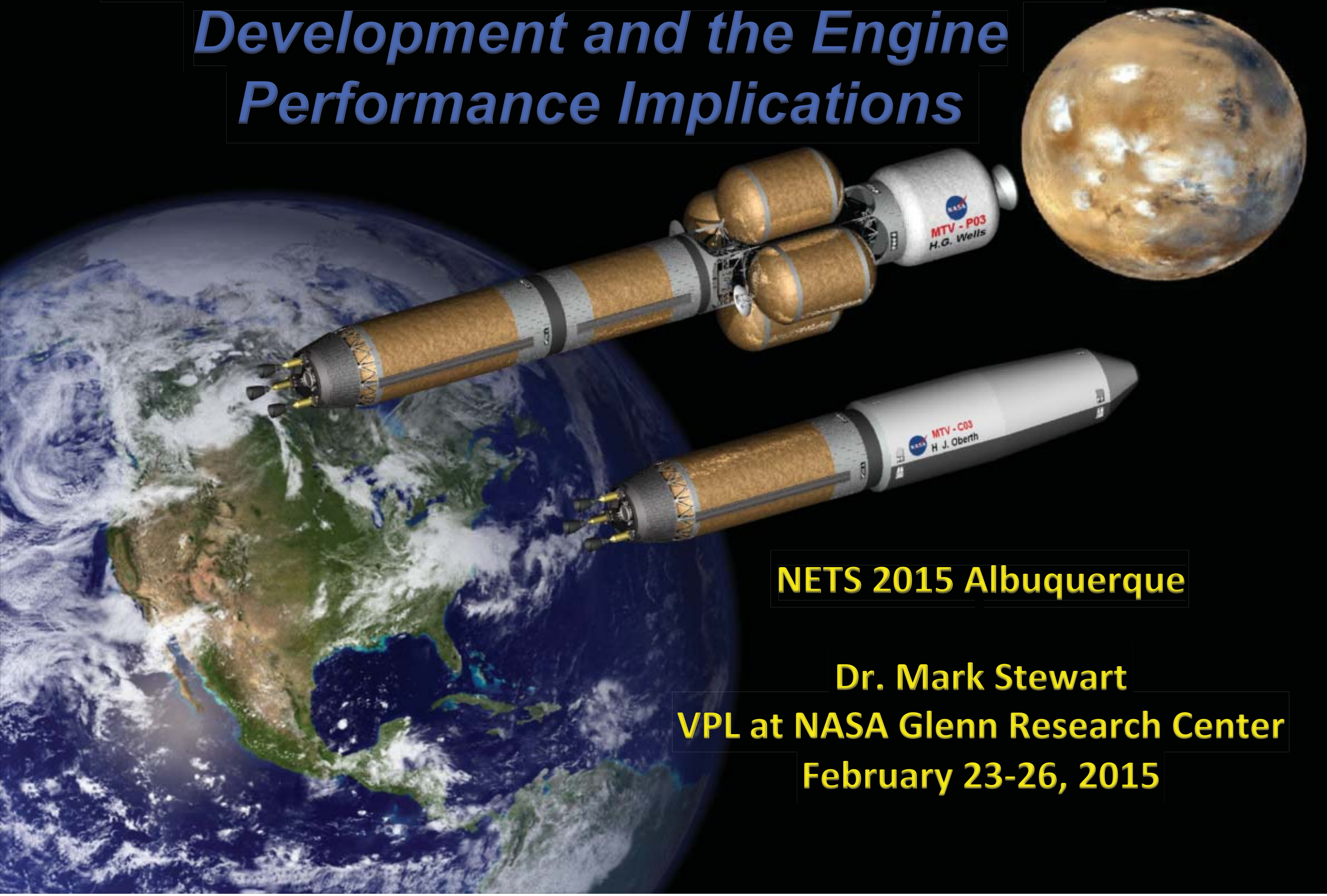
A Historical Review of Cermet Fuel Development and the Engine Performance Implications



NETS 2015 Albuquerque

Dr. Mark Stewart
VPL at NASA Glenn Research Center

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Outline

- Brief History
- Cermet sample testing during the NERVA/Rover era
  - Matrix/chart of samples tested & results
  - Comparison of approaches
- Important properties in context
  - Melting temperature
  - Vaporization rate
  - Chemical stability
- Engine performance
  - Location peak temperature
  - Heat deposition rate

History of Cermet Sample Testing

- 1949- NEPA investigated $Mo\text{-}UO_2$ and $W\text{-}UO_2$
- 1950's- Some further work
- 1961- Kennedy: "accelerate development of the **Rover nuclear rocket**"
- 1961- GE high-temperature materials program (HTMP & GE 710)
- 1962- Nuclear Propulsion Conference
  - LANL, LeRC, (GE) reported extensive testing results
  - UO_2 vaporization significantly reduced by thin tungsten cladding
  - UO_{2-x} reduction issue, uranium hydride formation, and sample cracking
- 1960's- DOE's ANL, Pacific Northwest labs
- 1968- ANL 200/2000 engine design, 2500^{o}C, <1% fuel loss, 10h, 25X
- 1968- Tighter budgets, terminal cermet fuel reports
- 1970~ Space race won: cancelled Apollo 18-20, manned Mars plans
- 1972- Rover/NERVA program cancelled
- Other cermet summaries: Haertling & Hanrahan, Lundberg & Hobbins

Performance of Historical Cermet Samples

A Broad Brush Painting of W-UO_2 Results

Unstable: Cracks or Forms Powder

Stable: Mass Loss > 5 %

Stable: Mass Loss < 5 %

Sample Group		1	2	3	4	5	6	7	8	9	10	11	12	13	14
UO_2 Only		✔						✔							
W-UO_2			✔	✔	✔	✔	✔		✔	✔	✔	✔	✔	✔	✔
Partial Clad (Not Edges)									✔			✔			
Full Clad										✔	✔		✔	✔	✔
Coated Fuel Particles				✔									✔		
Stabilizers (Various)							✔	✔	✔	✔		✔	✔	✔	✔
Temperature															
(C)	(K)														
2000	2273														
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- Considerable amount of cermet materials research in the early 1960's
- Over 200 W-UO_2 samples from five different labs: ANL, GE, LANL, LeRC, PNWL.
- Successes: full cladding, chemical stabilizers, coated particles
- Process improvement was done

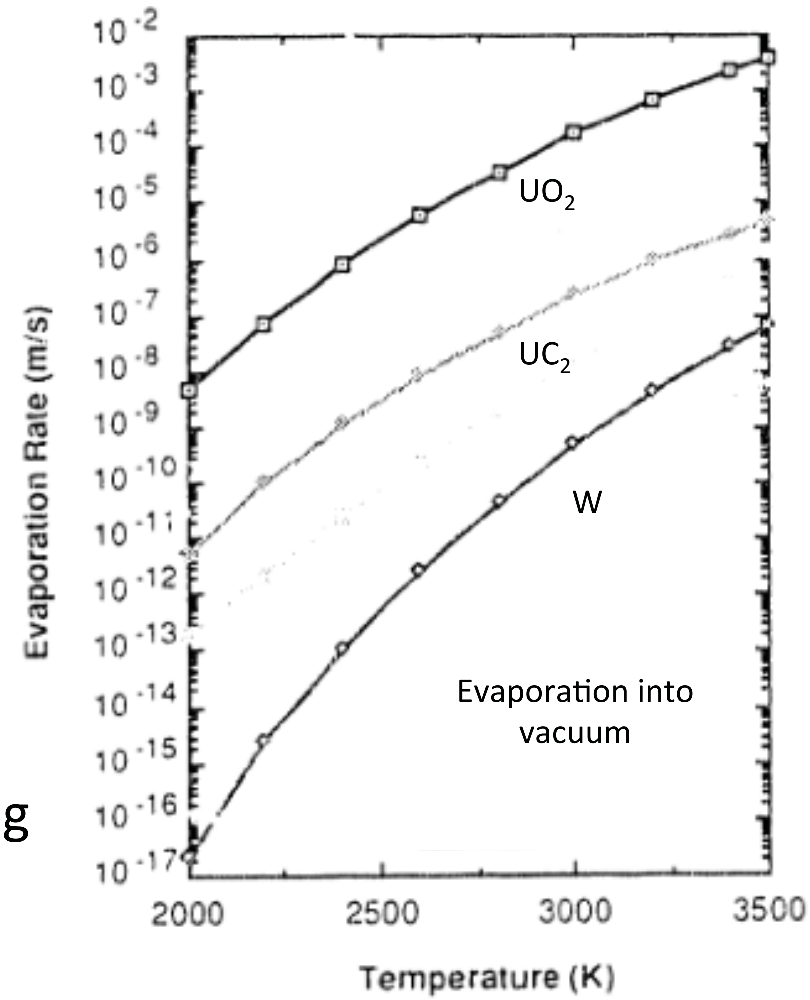
Melting Points & Vaporization Rates of NTP Reactor Fuels/Materials

Material/ Element	Melting Temperature (K)	Surface Vaporization Rate at 2800°K (mil/hr)
Tungsten	3680	< 0.01
Rhenium, Re	3453	0.1
Graphite	3915 (sublimes)	10
ZrC	3805	>>10
Tantalum Carbide, TaC	4150	0.1
Uranium Dioxide, UO_2	3075	6×10^3
Uranium Nitrides	Chemically Unstable	
Uranium Carbide, UC_2 NERVA Peewee	2835	10
UC-40 ZrC NERVA Composite	3050	2

Fuel Vaporization and Reactions

Coating/Cladding Needed

- Fuel vaporization is very high above 2000 K
- Cladding/particle coating needed
- At 1962 Nuclear Propulsion Conference
  - LANL (Lenz & Mundinger [9]): thin tungsten coatings reduce vaporization
  - LeRC (Saunders et al[13], McDonald[12]: fuel vaporization reduced 10X by cladding
- Face cladding is insufficient
  - Gluyas et al [13] demonstrated the need for full cladding

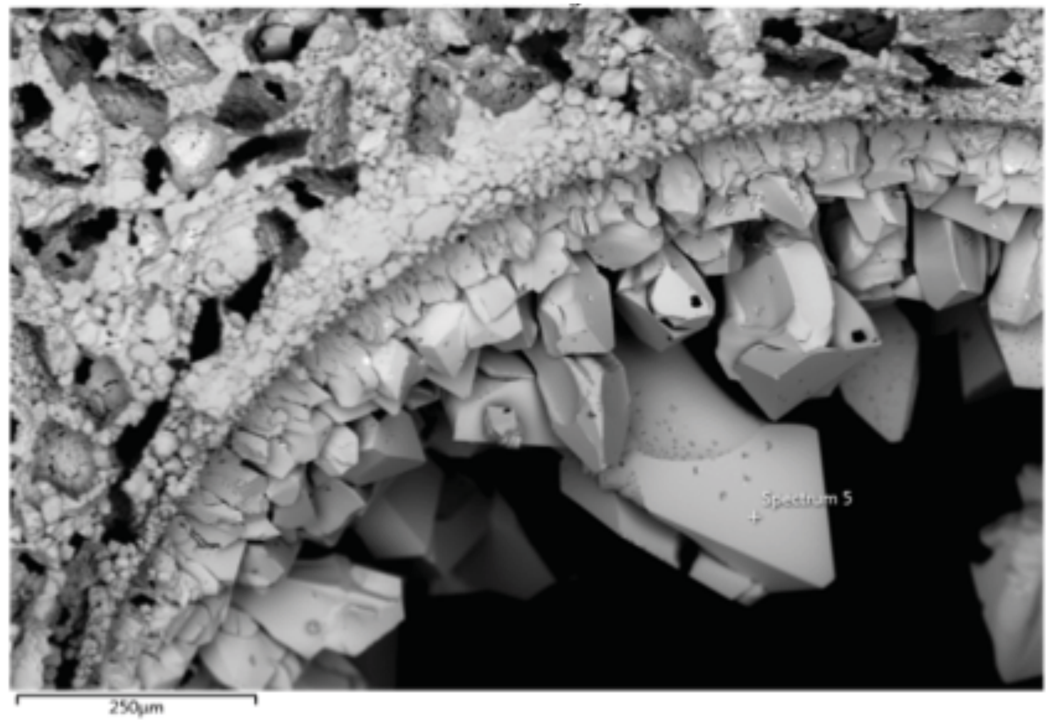


Evaporation rates of nuclear fuels and materials normalized to surface regression rates.

Lundberg, Hobbins EGG-M—92067

Fuel Vaporization Above 2000K

- Sample at 1900K for 30 minutes without significant mass loss
- At 2500K, sample experienced fuel evaporation



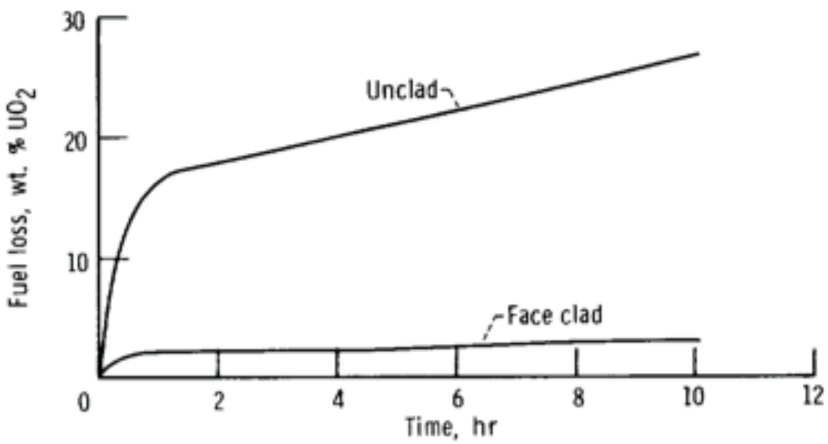


Figure 30. - Fuel loss as function of time at 2500° C in hydrogen for unclad and partially clad W - 20-volume-percent-UO_2 composites.

NASA TM X-1421

Performance of Historical Cermet Samples

W-UO_2 Results

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Stable: Mass Loss < 5 %

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Beals et al

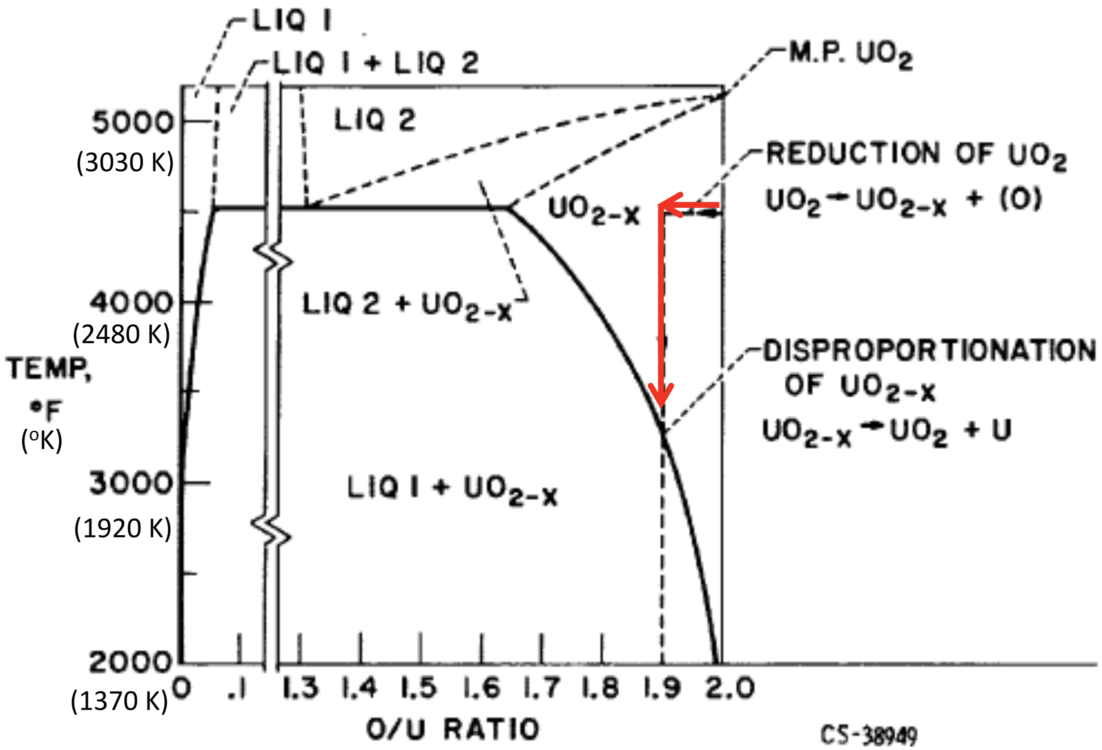
Baker et al

Beals et al

Gluyas, Gedwill

Lenz, Mundinger

High Temperature Behavior of UO_2



U-O-U-O-U-O-U-O-
I I I I
O O O O
I I I I
U-O-U-O-U-O-U-O-

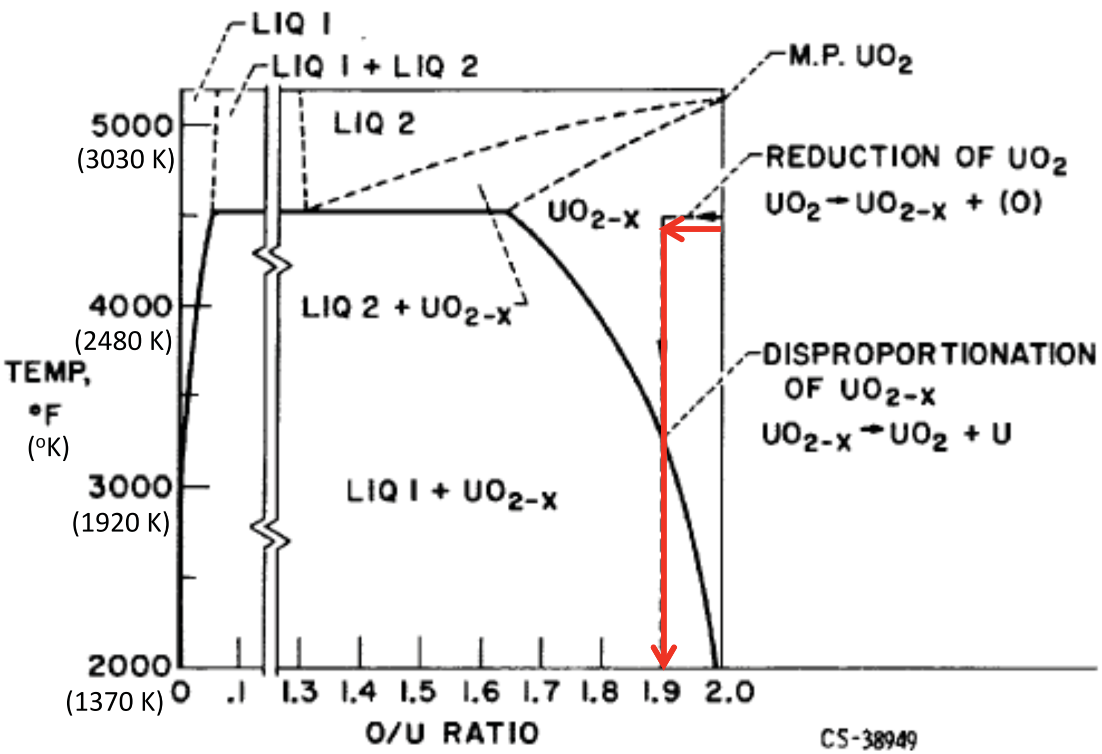
UO_2 structure
Idealized

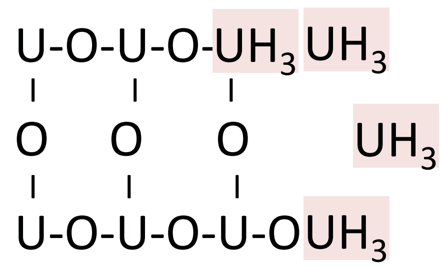
At temperatures above 2000°K, UO_2 becomes deficient in oxygen.
With cooling, free uranium forms.
Stabilizers (Gd_2O_3, Y_2O_3) interfere with this reduction.

Stability of UO_2, and Chemical Stabilizers

- 1960 Anderson: UO_2 reduces to UO_{2-x}
- 1962 LeRC, LANL, GE: UO_{2-x}, free uranium & UH_3 forms, sample cracking
- 1965 Beals, et al: Hydride, UH_3, formation is accompanied by a disruptive volume change that destroys the integrity of the specimen
  - UO_2 was "heated to 2300 C in flowing dry hydrogen for 10 min. When cooled in hydrogen to below 500 C, the pellet disintegrated with sufficient force to shatter the (foil) crucible. The residue was a very fine black powder."
  - "crumbling or powdering of the specimen"
  - UH_3 forms between 370 K and 620 K; can cool in non-hydrogen environment
- Addition of rare-earth oxides improves stability, particularly gadolinium Gd_2O_3, also Y_2O_3
  - UO_2 at 2570 K crumbles
  - UO_2 10 mol% Gd_2O_3 heated to 2770K had 6-12 % weight loss
  - At 2920 K, needed 5 mol% $GdO_{1.5}$ + 5 mol% $FeO_{1.5}$ for stability

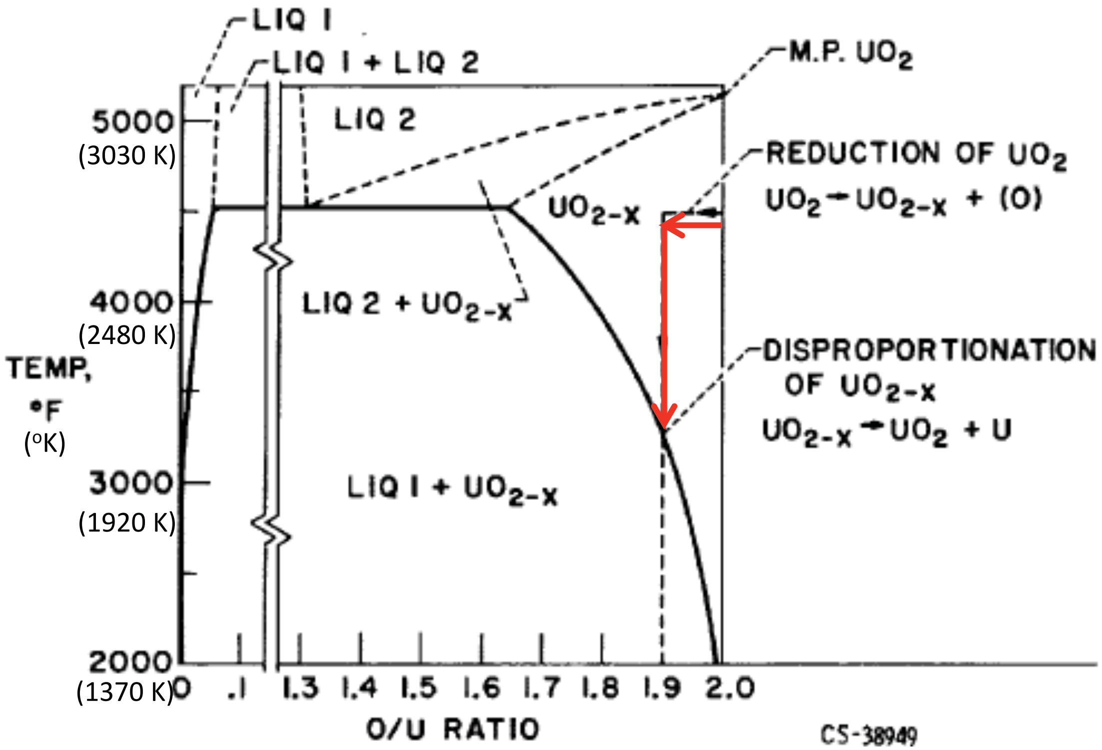
High Temperature Behavior of UO_2



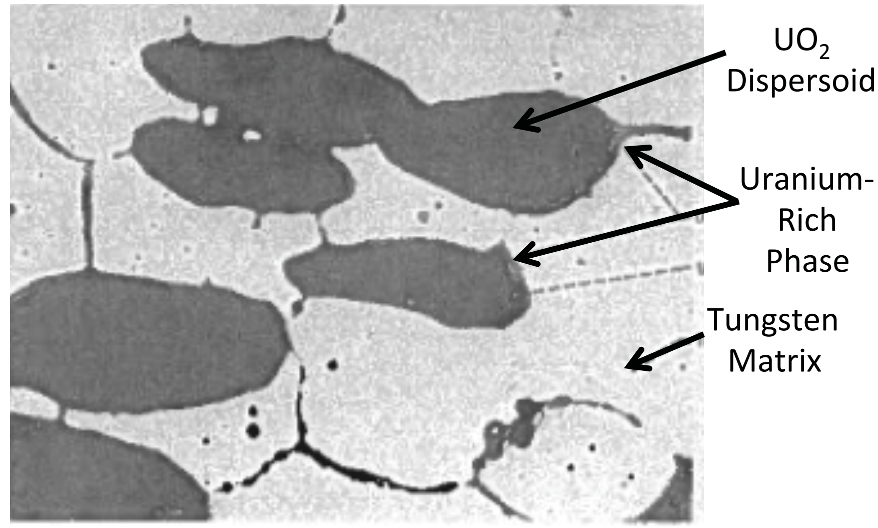


With cooling below 2000°K, free uranium forms.
Below 770°K, free uranium combines with hydrogen to form uranium hydride, UH_3.
This is hydrogen embrittlement

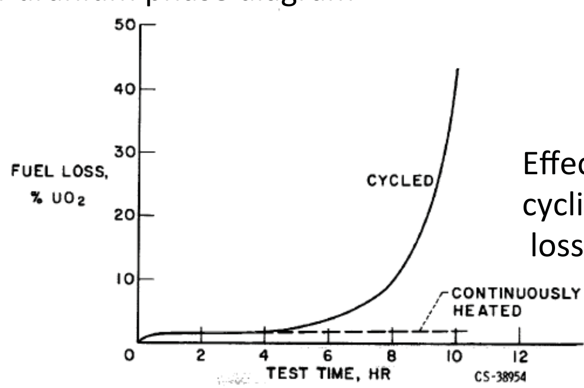
Chemical Stability of UO_2 with Thermal Cycling



Oxygen-uranium phase diagram



Micrograph of thermally cycled W-20 vol% UO_2 cermet showing free U at grain boundaries. The specimen was heat treated for five 1-h intervals at 2770 K in H_2 with cooling to room temperature between cycles.



Effect of thermal cycling on fuel loss at 2750 K.

NASA X66 51413
NASA TM-X-1421

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Beals et al

Beals et al

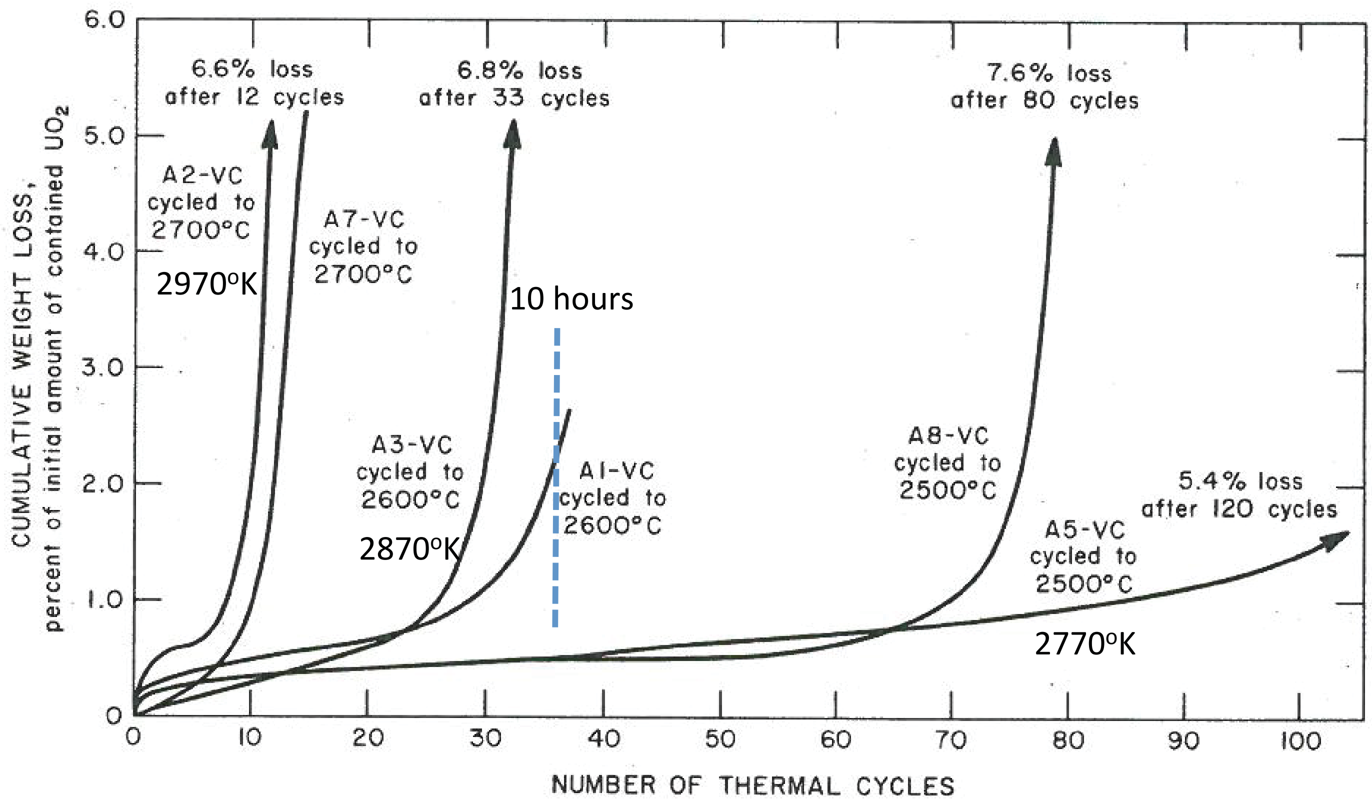
Gluyas, Gedwill

ANL

Pattern of Performance in Cermet Fuel Samples

Thermally cycled in furnace—not rocket/reactor conditions

- Six samples
- Low-pressure hydrogen,
- ANL-7150 suggests the hydrogen is static, or nearly so.
- Testing with flowing hydrogen at engine pressures would reduce performance.
- Comparable results found at 2500°C in reference 13, Y_2O_3, 20-35v% UO_2, flowing hydrogen
- Each 100°C increase in temperature significantly decreases lifetime



Fuel loss behaviors of tungsten-clad W-66 v/o (10m/o GdO1.5-stabilized UO_2) cermet samples (not fuel elements) thermally cycled to 2770 K, 2870 K, and to 2970 K.

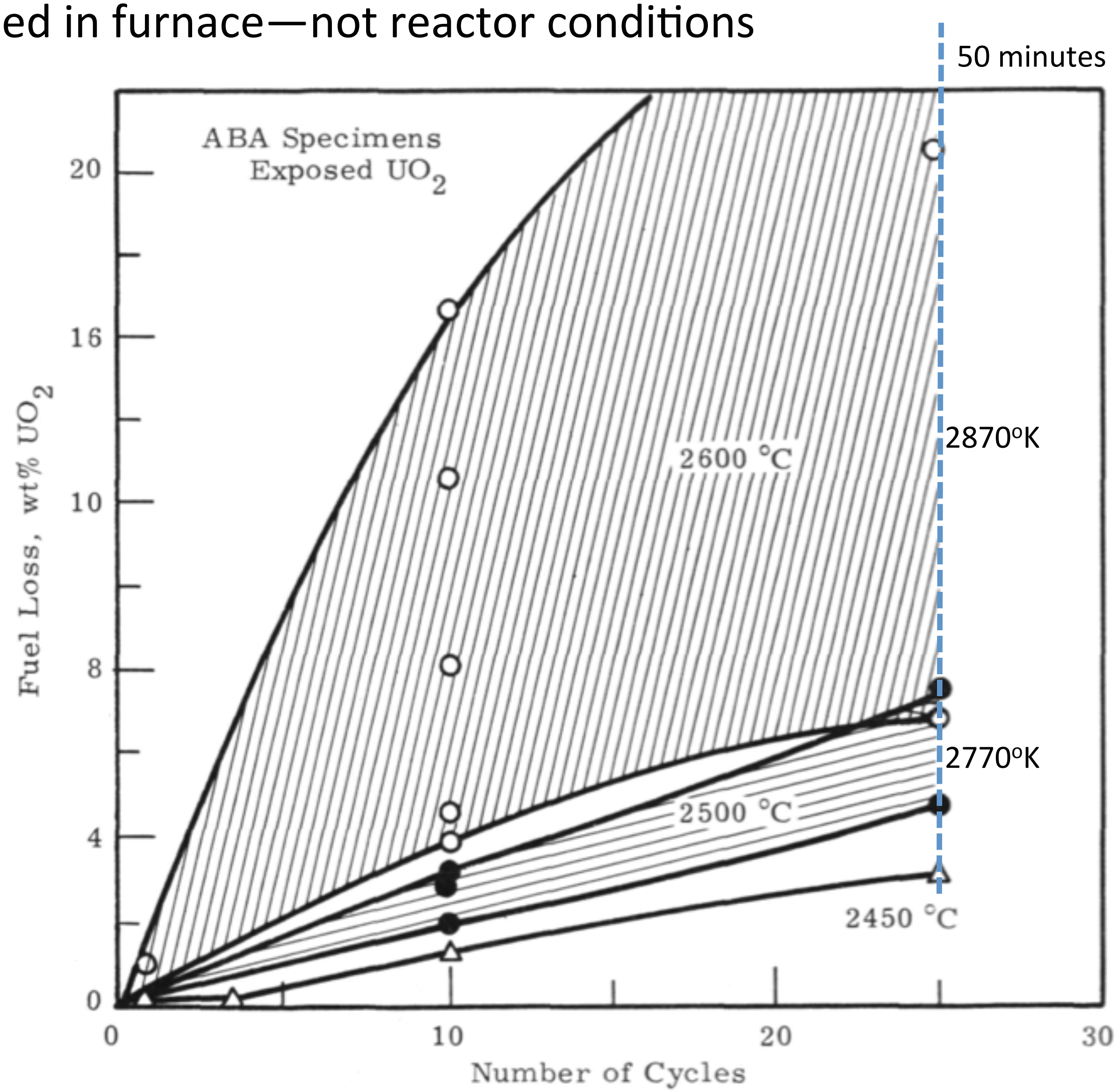
ANL-7150

Pattern of Performance in Cermet Fuel Samples

Thermally cycled in furnace—not reactor conditions

- Fuel particles coated with W, no cladding
- High-pressure, static hydrogen,

Thermal Cycling Behavior of Tungsten-Coated UO_2-W (13.3 vol% UO_2) Cermets Under Accelerated Test Conditions in 68 atm Static Hydrogen.

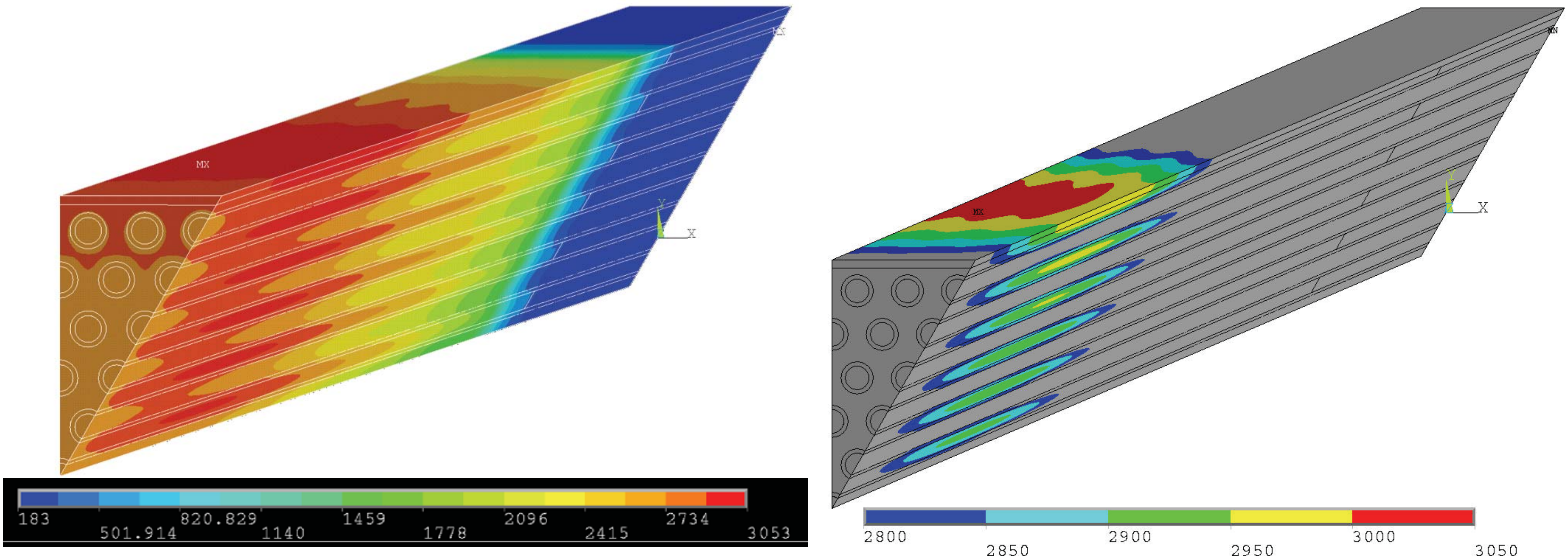


NASA CR-54840, p. 48

What Does a Fuel Element Designer Do with Material Performance Data?

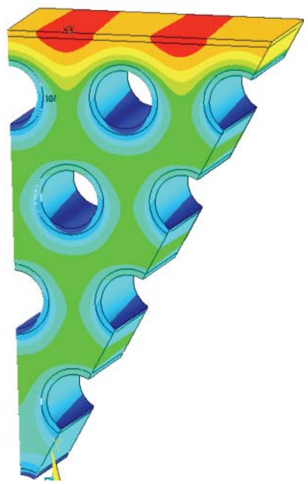
- Engine/Fuel designer must:
  - Highest possible propellant outflow temperature
  - Minimum peak fuel temperature
  - Nuclear criticality & control
  - Engine system performance (turbopump, nozzle)
  - Acceptable fuel loss, maintain fuel integrity
- High fidelity simulations help understanding
  - Neutronics simulations predict criticality
  - High-Fidelity fluid / thermal / structural simulations
  - Can simulate materials and performance

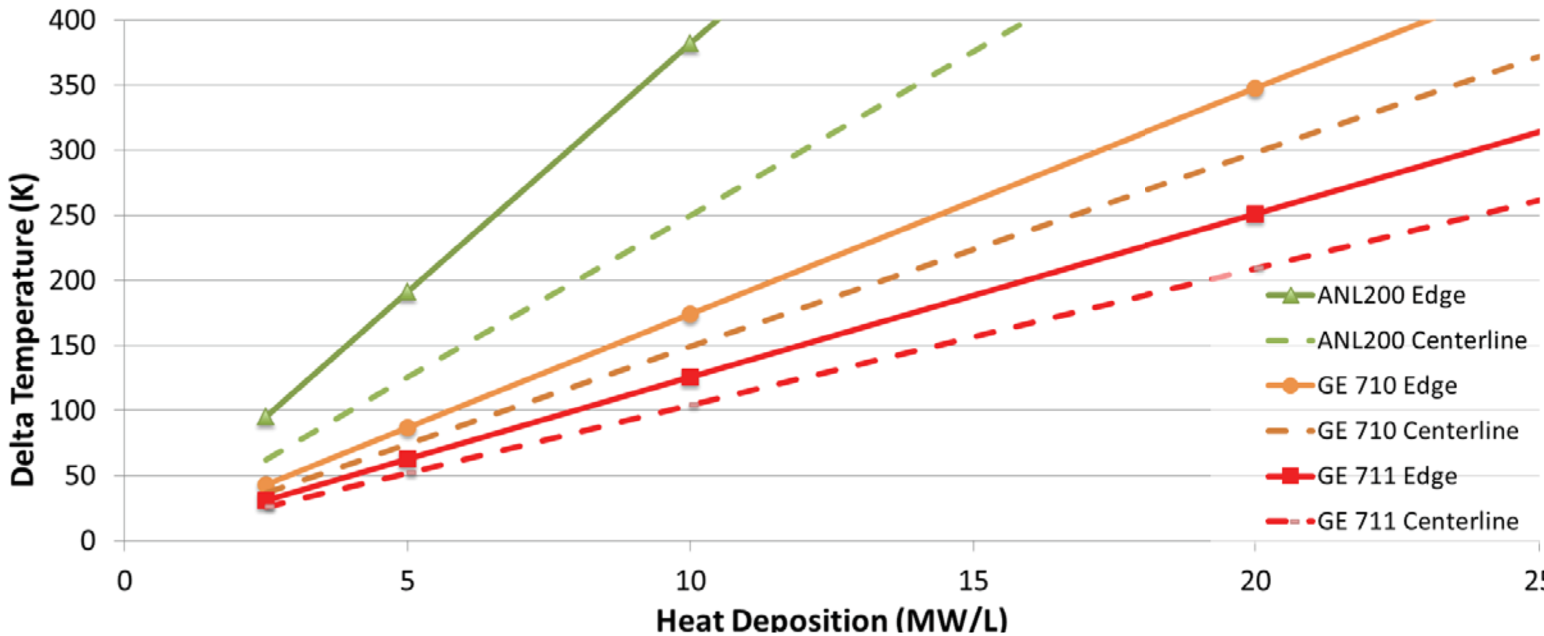
Where is the Hottest Cermet Fuel?



Predicted temperature distribution through a GE 711 cermet fuel element (left) and detail of the hottest 250K region of the fuel element (right).

Fuel Temperature Differences Versus Heat Deposition Rate



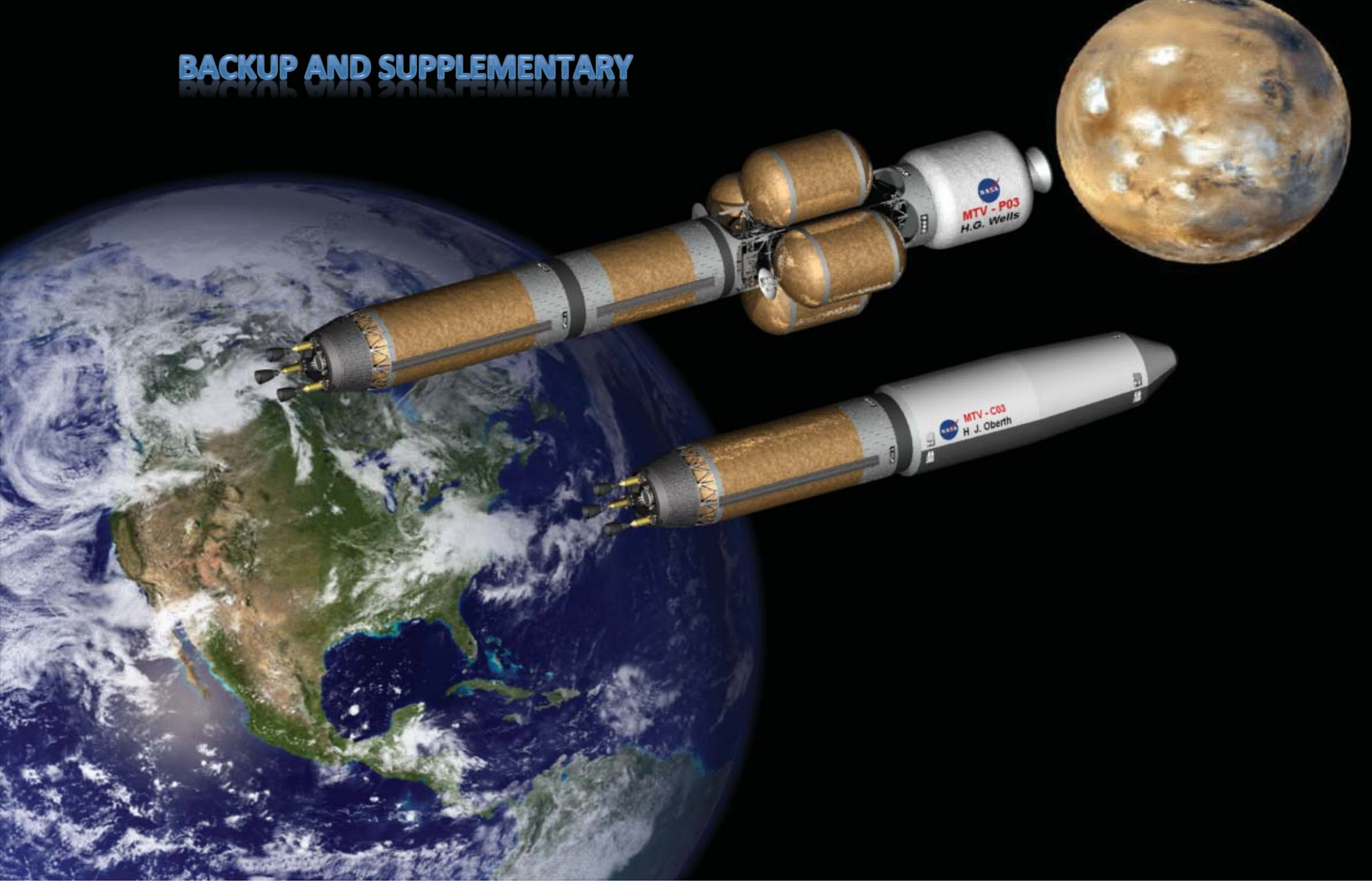


Predicted temperature difference, fuel peak at edge to coolant channel (solid) and fuel centerline to coolant channel (dashed) for several cermet fuel geometries.

Summary and Conclusions

- To better understand Cermet engine performance, examined historical material development reports
- Two issues:
  - High vaporization rate of UO_2
  - High temperature chemical stability of UO_2
- Cladding and chemical stabilizers each result in large, order of magnitude improvements in high temperature performance
- Some long duration, low mass-loss, samples at 2770^{o}K
- Few samples were tested above 2770^{o}K
- Contemporary testing may clarify performance

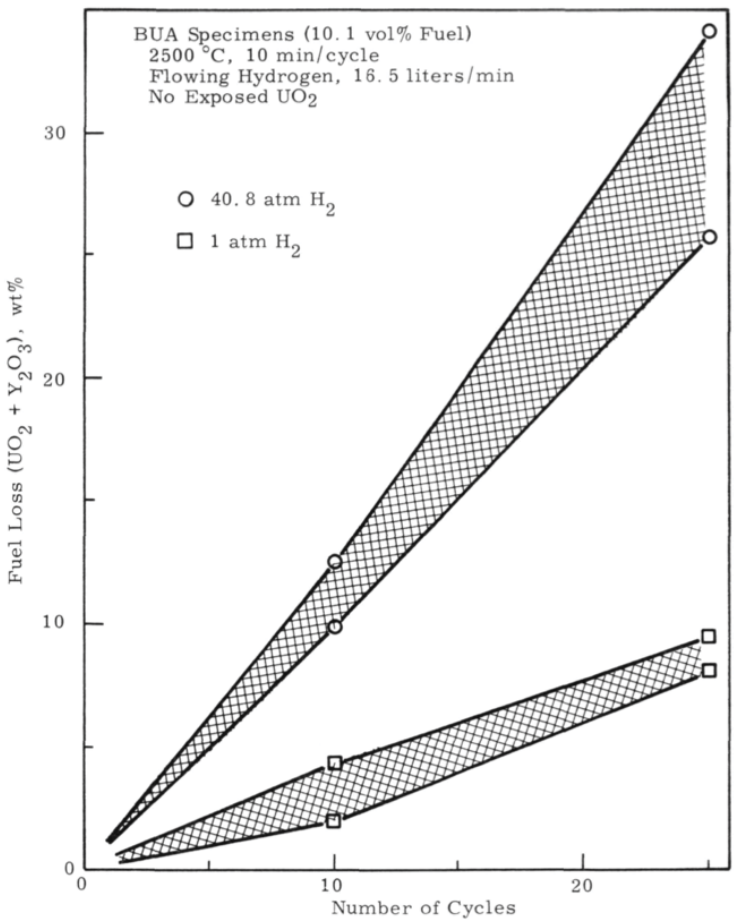
BACKUP AND SUPPLEMENTARY



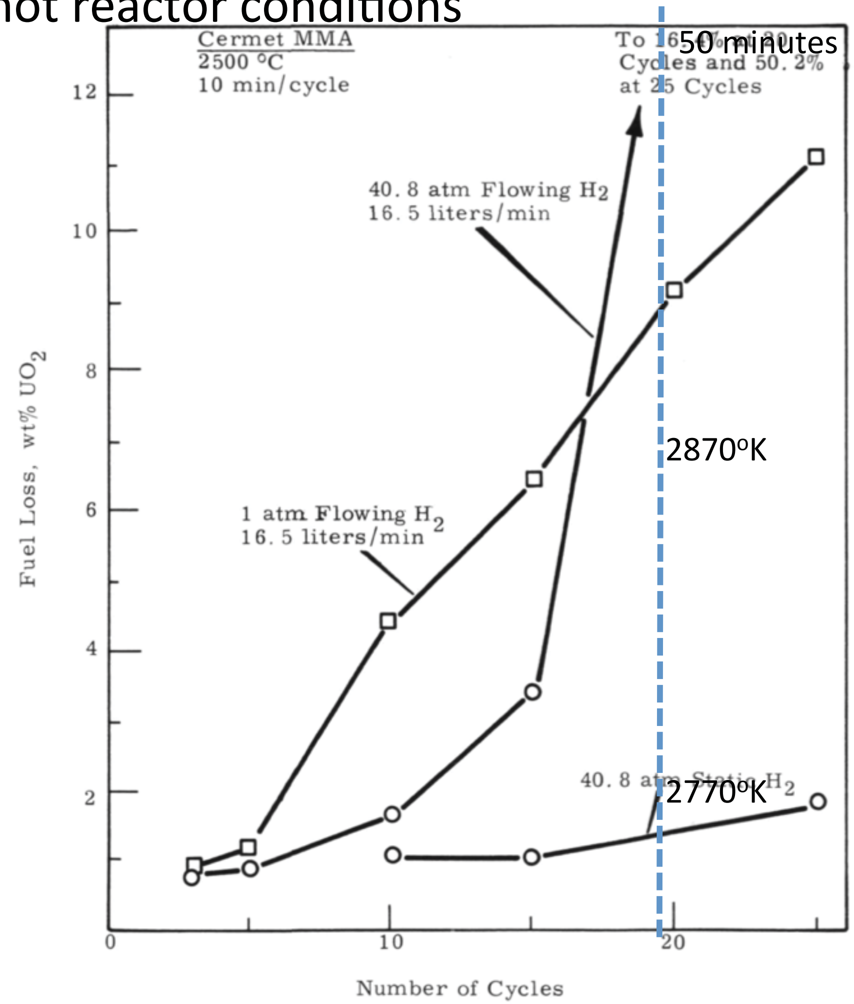
Pattern of Performance in Cermet Fuel Samples

Thermally cycled in furnace—not reactor conditions

- High-



Thermal Cycling Behavior of W-UO_2 Coated Particle Cermets Containing 10 Mole% Y_2O_3 in UO_2 Solid Solution.



Effect of Pressure and Flow Rate on the Thermal Cycling Behavior of a Tungsten-Coated UO_2-W Cermet Containing 13.3 vol% UO_2.

NASA CR-54840, p. 48

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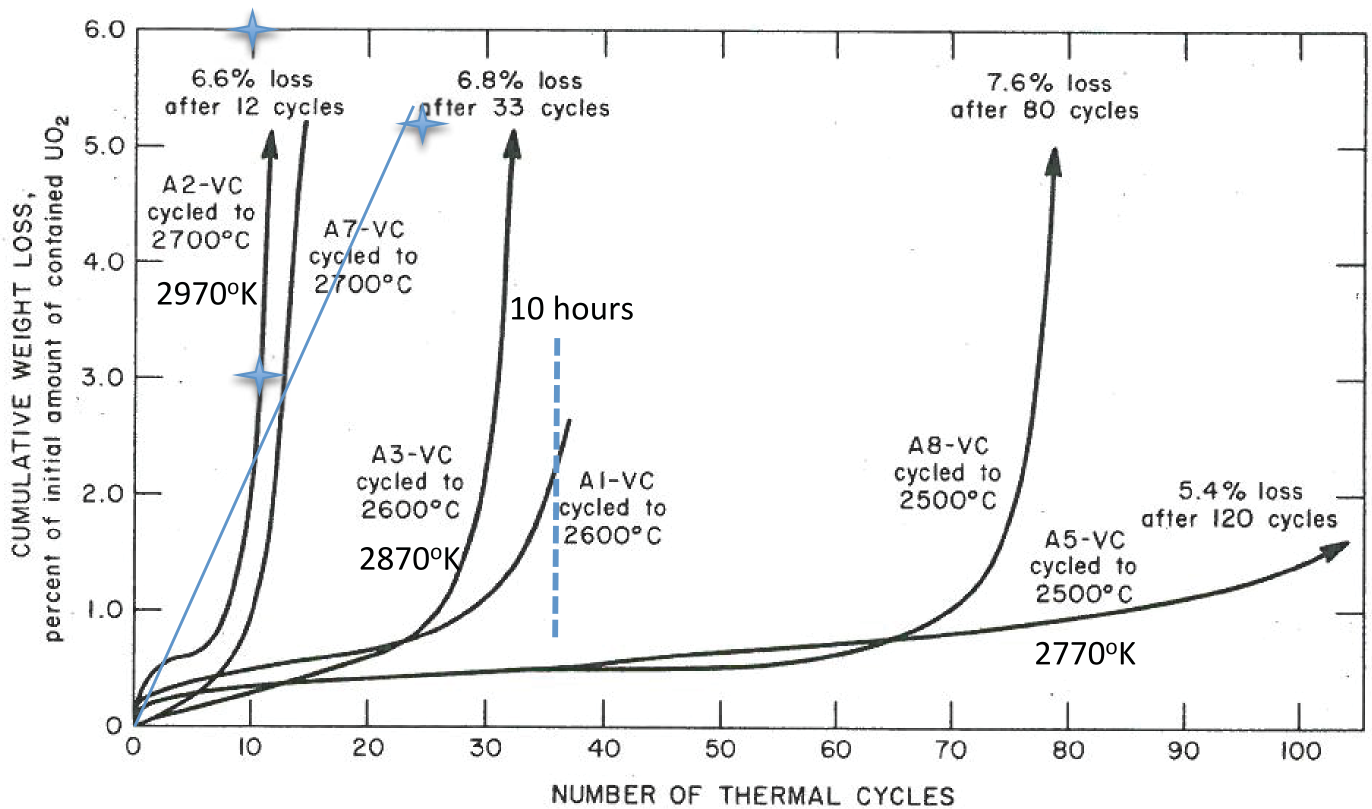
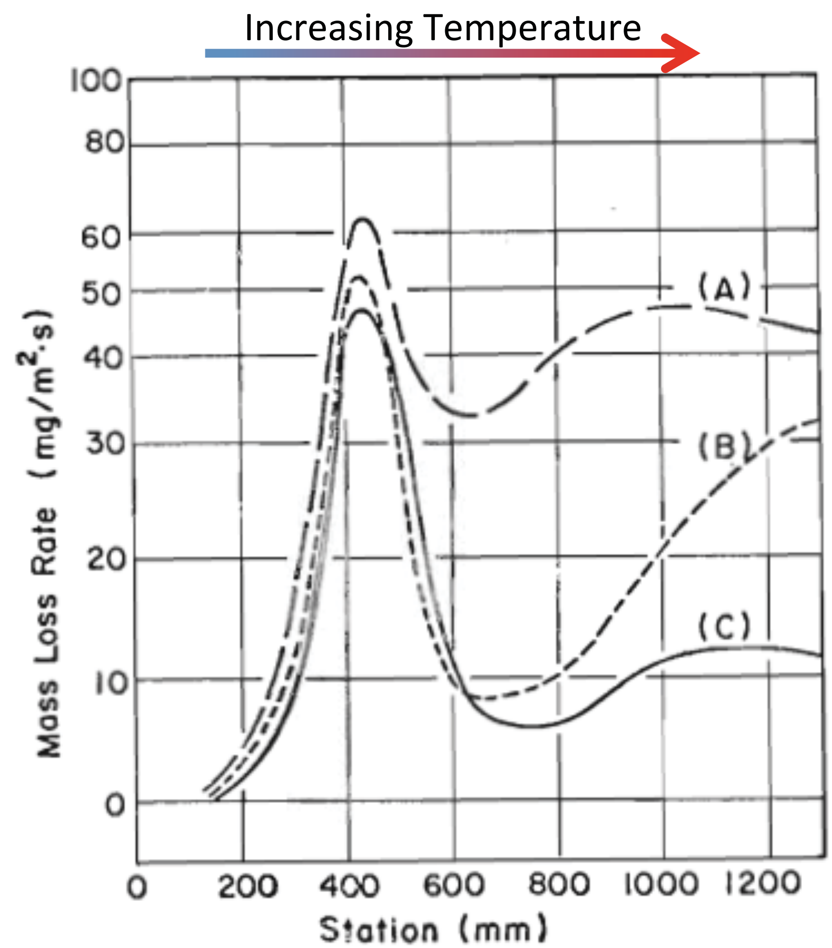


Fuel loss behaviors of tungsten-clad W-66 v/o (10m/o GdO1.5-stabilized UO_2) cermet samples (not fuel elements) thermally cycled to 2770 K, 2870 K, and to 2970 K.

ANL-7150

Mid-Band Erosion in NERVA Composite Fuel Elements

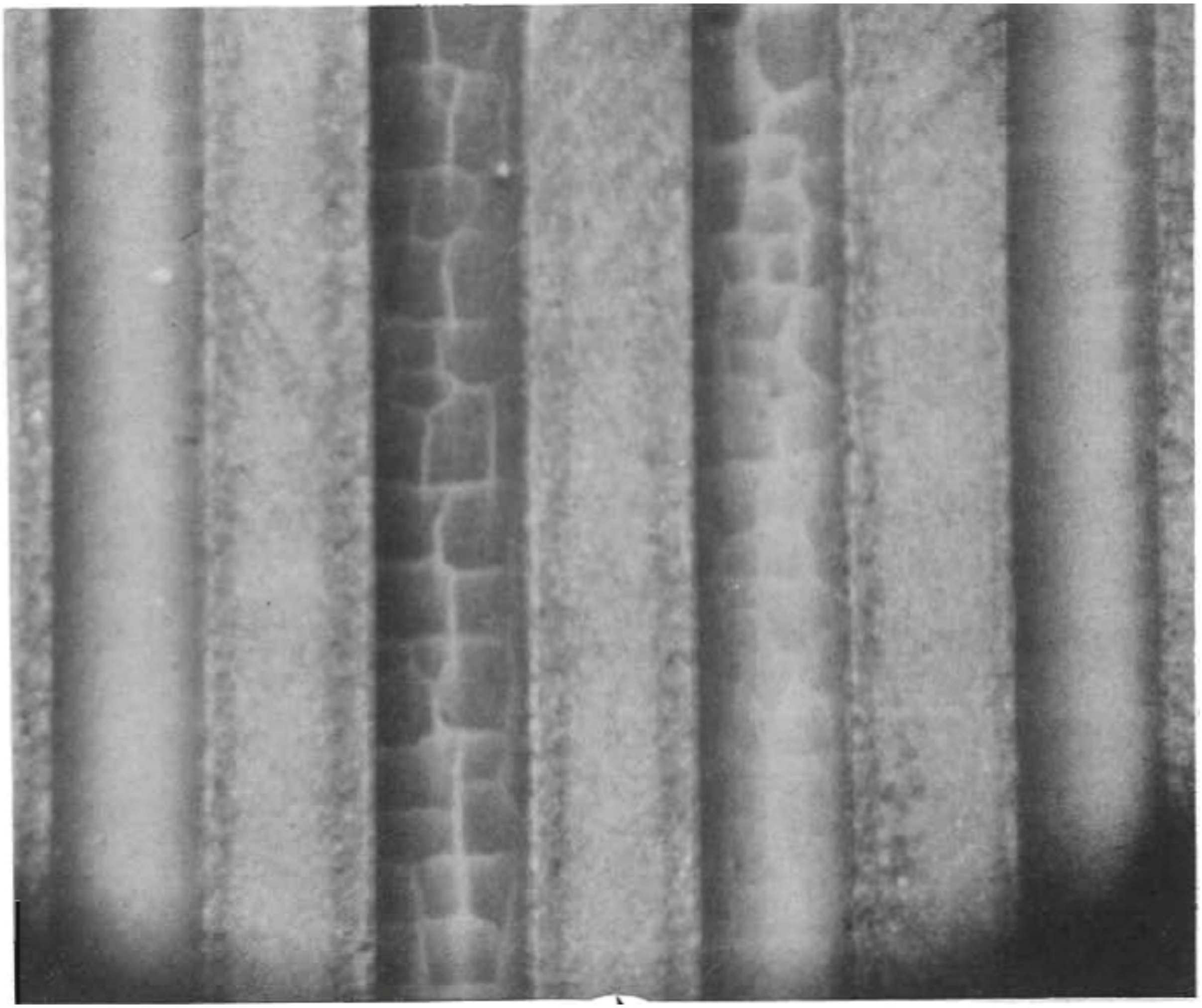
- High CTE composite fuel elements were crack-free as fabricated
- Hot end fuel loss agreed with predictions
- Midrange losses were unexpected, 2/3 of total
- Cold end coating cracks caused midrange loss, $H_2 + C \rightarrow CH_3$ & C_2H_2



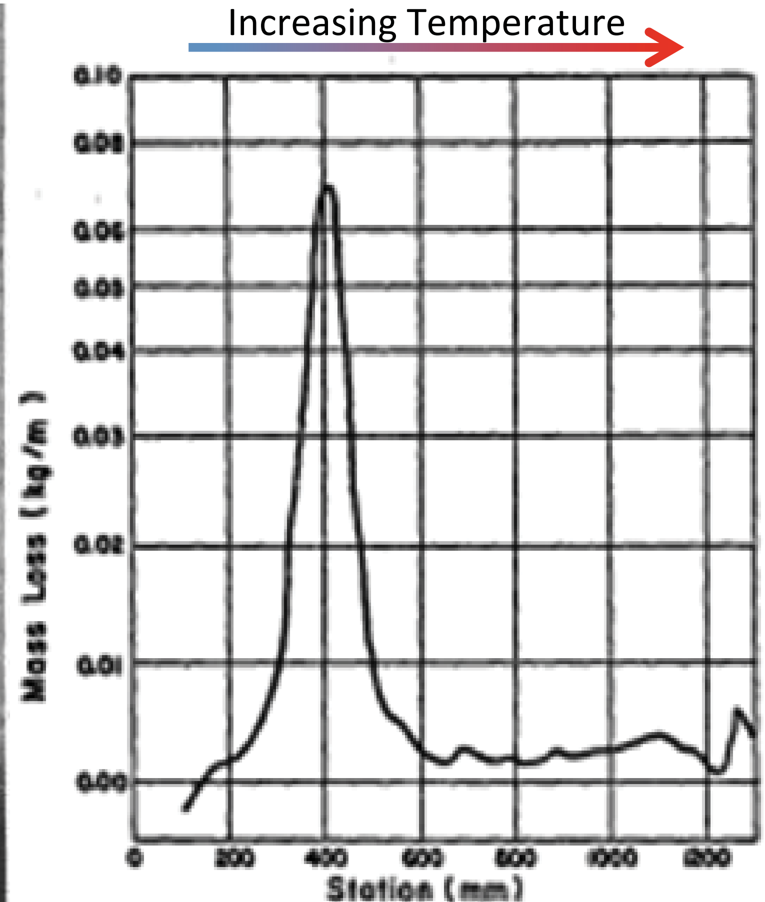
Mass loss rates per unit surface area of coolant channel versus station for graphite and composite elements. (A) average for 102 Pewee-1 graphite fuel elements coated with NbC, (B) average for 12 Pewee-1 graphite fuel elements coated with ZrC, (C) average for 23 NF-1 high-CTE composite fuel elements coated with ZrC, adjusted to the Pewee-1 test temperature.

LA-5398-MS

Pattern of Cracking / Erosion in NERVA Fuel



NERVA fuel element interior coolant channels experienced coating cracks in the NF-1 test, while edge channels retained their coatings. Mid-passage erosion region.

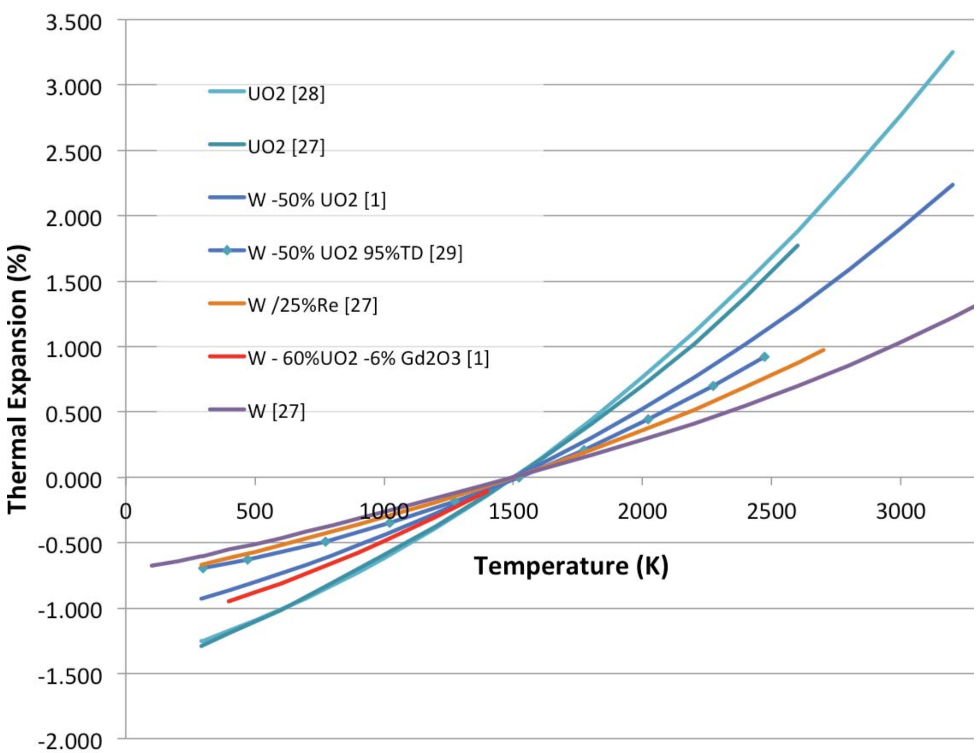


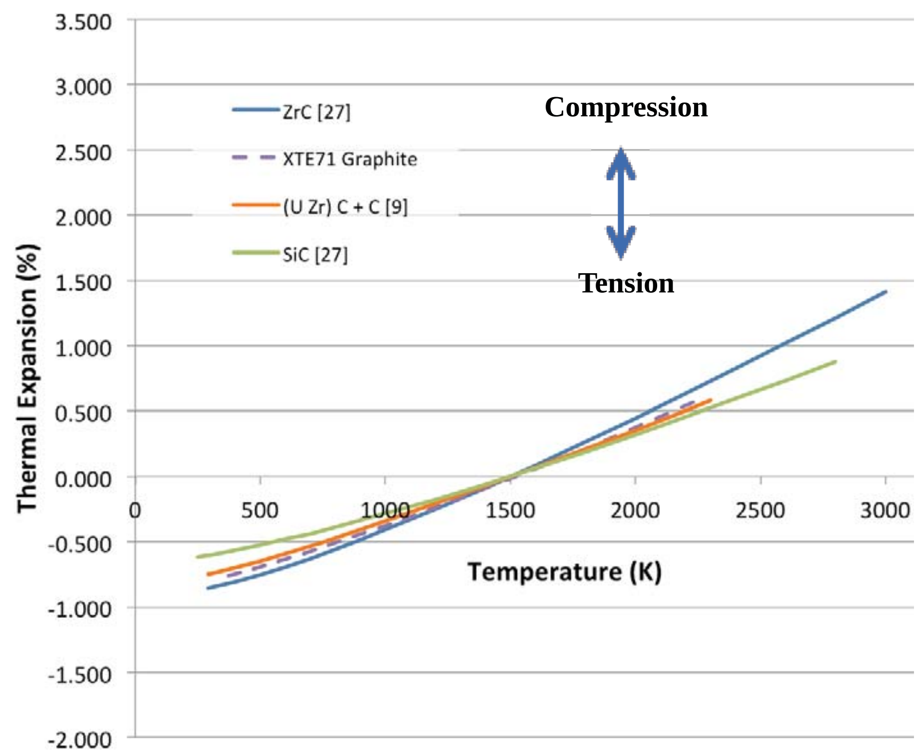
Mass loss versus length for (U,Zr)C-graphite fuel element in NF-1 test. CTE > 6.5 μ m/mK.

LA-5398-MS.

Stress Sources: Differential Thermal Expansion

Cermet and Graphite-Based Fuels





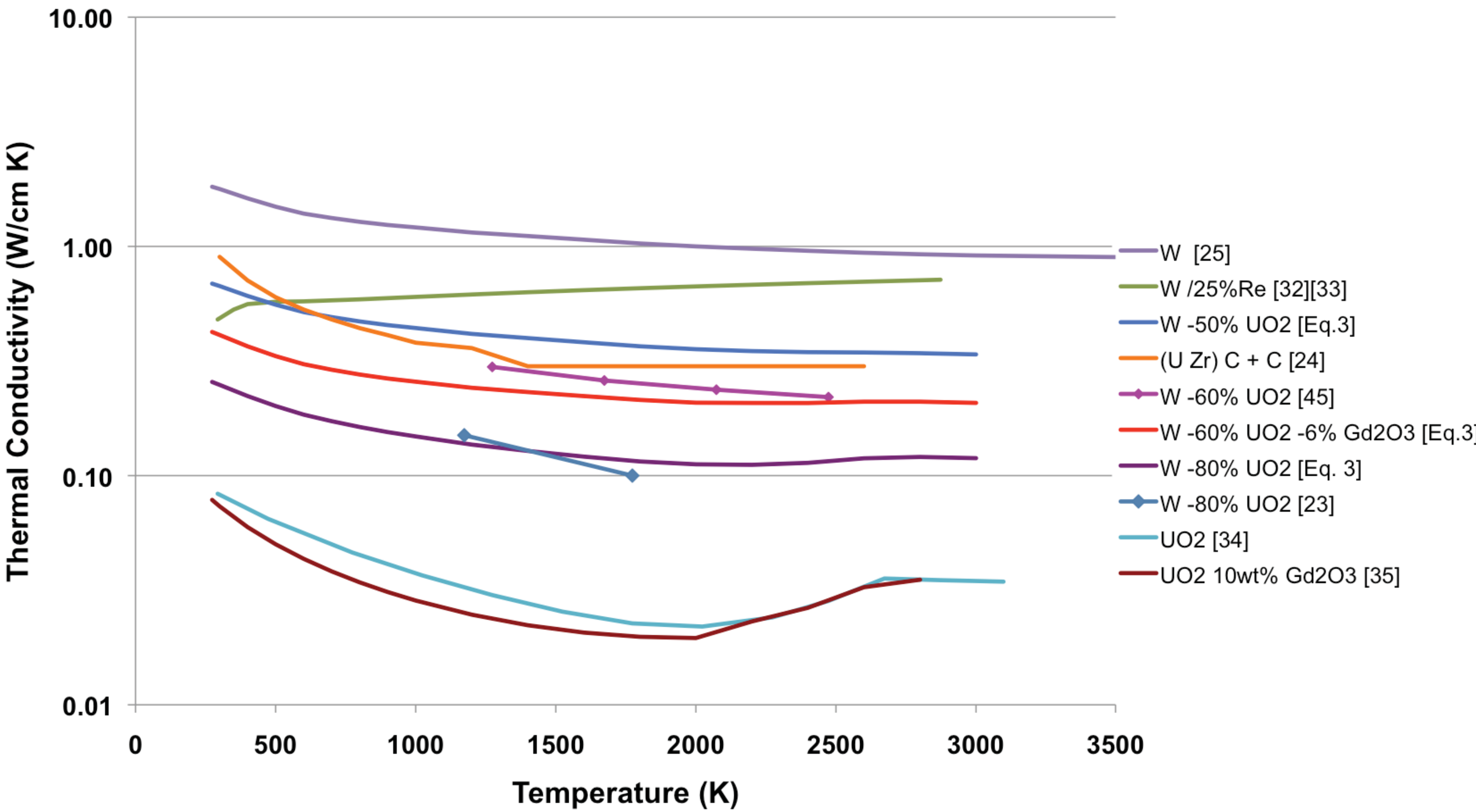
Cermet Fuels

Coatings in *compression* on cool down

Graphite-Based Fuels

Coatings in *tension* on cool down

NTP Fuel Elements: Thermal Conductivity



NTP Fuel Elements: Modulus of Elasticity